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For: Thixotropy-imparting agent

VERIFICATION OF TRANSLATION

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Date: December 19, 2005

Keiji Tokieda

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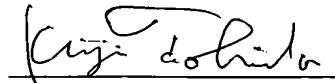
(Title of the Invention)
       Thixotropy-Imparting Agent

(Technical Field)
5   The present invention relates to a thixotropy-
imparting agent. More particularly, the invention
relates to a highly thixotropy-imparting agent
comprising chain clay mineral particles and having a
particularly large thixotropic index.
10   (Background Art)
         In the field of coating materials, it is a
requirement that a suppressed viscosity is exhibited
at the time of preparing the coating materials (at the
time of mixing the components) and at the time of
15   applying the coating materials from the standpoint of
workability. For this purpose, therefore, a
thixotropy-imparting agent is used to lower the
viscosity under the conditions of high shearing
forces. After having been applied, on the other hand,
20   a high viscosity must be exhibited to prevent dripping
calling for a high thixotropy with a large thixotropic
index (TI).
         The chain clay minerals (holmite) such as
sepiolite and attapulgite have a three-dimensional
25   chain structure like alternately piling talc bricks.
It has been known that holes formed among the chain
gaps possess large surface areas serving as a factor
that produce a high viscosity or thixotropy when the
chain clay mineral is obtained in a slurry form,
30   exhibiting, for example, thixotropy and rheopexy (see
JP-A-4-228461).
         However, the chain clay minerals such as the
sepiolite and the like that are naturally produced
contain impurities that differ depending upon the
35   region of production; i.e., the chain clay minerals do

not all have large thixotropic indexes (TI).
Therefore, it has been desired to provide an agent
capable of imparting a high degree of thixotropy
maintaining stability.

5   (Disclosure of the Invention)

It is, therefore, an object of the present
invention to provide a highly thixotropy-imparting
agent comprising chain clay mineral particles and
having a large thixotropic index.

10   According to the present invention, there is
provided a thixotropy-imparting agent comprising chain
clay mineral particles, characterized in that said
chain clay mineral particles has:

a thixotropic index (TI) defined by the following

15   formula,

$$TI = \eta_6 / \eta_{60}$$

wherein η_6 is a viscosity (at 25°C) of a
dispersion solution obtained by dispersing the
chain clay mineral particles in a predetermined

20   dispersion medium as measured at a rotational
speed of 6 rpm, and η_{60} is a viscosity (at 25°C)
of the above dispersion solution as measured at a
rotational speed of 60 rpm,

of not smaller than 4.0 in a dispersion solution

25   obtained by dispersing chain clay mineral particles at
a concentration of 7% by weight in diethylhexyl
phthalate (DOP) as a dispersing medium and not smaller
than 3.0 in a dispersion solution obtained by
dispersing chain clay mineral particles at a

30   concentration of 3% by weight in water as a dispersion
medium;

a bulk density of not larger than 0.125 g/ml; and
a particle size distribution of secondary
particles as measured by a laser method, in which

35   particle sizes of larger than 1.0 μm but not larger

than 30 μm are not less than 70% by weight and particle sizes of not larger than 1.0 μm are in a range of 5 to 30% by weight.

According to the present invention, there are further provided a coating material composition, an adhesive composition and a resin composition containing the above thixotropy-imparting agent.

In the thixotropy-imparting agent of the present invention, it is desired that:

1. the chain clay mineral particles have an average aspect ratio of 7.5 to 9.5;

2. the chain clay mineral particles are such that a primary particle shape thereof has an average fiber length of 0.45 to 0.80 μm as measured by using an electron microscope;

3. the chain clay mineral is holmite clay mineral;

4. the holmite clay mineral is sepiolite or attapulgite;

5. the holmite clay mineral is sepiolite, and when the peak height stemming from the plane (110) of sepiolite is regarded to be 100%, the peaks stemming from dolomite and calcite have intensity ratios of peaks of not larger than 25% in the X-ray diffraction measurement;

6. the holmite clay mineral is attapulgite, and when the peak height stemming from the plane (110) of attapulgite is regarded to be 100%, the peak stemming from calcite has an intensity ratio of peak of not larger than 50%.

According to the present invention, a high thixotropy having a large thixotropic index can be imparted to the coating materials by using particular chain clay mineral particles.

(Brief Description of the Drawings)

Fig. 1 is a diagram showing X-ray diffraction

images of sepiolites of Example 1 and of Comparative Examples 3 and 4;

Fig. 2 is a diagram showing particle size distributions of secondary particles of a thixotropy-imparting agent of Example 1;

Fig. 3 is a scanning electron microphotograph (SEM) of the thixotropy-imparting agent of Example 1 (magnification: 20,000 times);

Fig. 4 is a scanning electron microphotograph (SEM) of a sample of Comparative Example 4 (magnification: 20,000 times); and

Fig. 5 is a diagram showing X-ray diffraction images of attapulgites of Example 3 and of Comparative Example 5.

(Best Mode for Carrying Out the Invention)

(Chain clay minerals)

If briefly described, it is important in the present invention to use chain clay mineral particles having a particular primary particle shape and a particle size distribution. More preferably, the present invention is based on a novel discovery that by using chain clay mineral particles having a particular X-ray diffraction peak and by adjusting the particle size, a high degree of thixotropy can be imparted as compared to that of using the conventional agents.

Representative examples of the chain clay minerals used in the present invention include sepiolite, attapulgite and palygorskite which are holmite clay minerals. These chain clay minerals are the hydrated silicates of magnesium. For example, sepiolite is generally produced in the serpentine as a secondary mineral, and attapulgite is produced being altered from the augite and hornblende in the aqueous rock. Attapulgite is often called palygorskite

depending upon the use or the region of production.

Sepiolite ideally has a chemical structure represented by the following formula (I),

$$(OH_2)_4(OH)_4Mg_8Si_{12}O_{30} \cdot 6\text{-}8\ H_2O \quad \text{--- (I)}$$
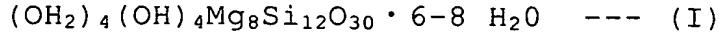
and, when generally calculated, has the following representative composition (when dried at 110°C for 2 hours).

General chemical composition of sepiolite:

SiO_2	52.50 (% by weight)
MgO	22.8
Al_2O_3	1.7
Fe_2O_3	0.8
CaO	0.8
H_2O^+	10.5
H_2O^-	11.0

Ideally, further, attapulgite has a chemical structure represented by the following formula (II),

$$(OH_2)_4(OH)_2Mg_5Si_8O_{20} \cdot 4H_2O \quad \text{--- (II)}$$
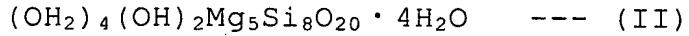
and, when generally calculated, has the following chemical composition (when dried at 110°C for 2 hours).

General chemical composition of attapulgite:

SiO_2	57.6 (% by weight)
MgO	22.3
Al_2O_3	0.9
Fe_2O_3	0.8
CaO	0.5
H_2O^+	9.9
H_2O^-	8.0

In the present invention, it is particularly desired to use sepiolite among the above chain clay minerals. That is, the thixotropy can be evaluated in terms of the thixotropic index (TI) defined by the following formula,

$$TI = \eta_6 / \eta_{60}$$

wherein η_6 is a viscosity (at 25°C) of a

dispersion solution obtained by dispersing the above chain clay mineral particles in a predetermined dispersion medium as measured at a rotational speed of 6 rpm, and η_{60} is a viscosity (at 25°C) of the above dispersion solution as measured at a rotational speed of 60 rpm, and the higher the thixotropic index (TI), the higher the thixotropy. Sepiolite has a thixotropic index (TI) of not smaller than 4.0 and, preferably, not smaller than 4.2 in a dispersion solution (clay mineral particle concentration of 70% by weight) using diethylhexyl phthalate (DOP) as a dispersion medium and a thixotopic index (TI) of not smaller than 3.0, preferably, not smaller than 5.0 and, more preferably, not smaller than 7.5 in the water which is used as the dispersion medium, exhibiting a very high thixotropy when blended in either the organic dispersion medium or the water dispersion medium. When compared with sepiolite, on the other hand, attapulgite, usually, exhibits a low thixotropic index (TI) and, particularly, a low thixotropic index in the water dispersion medium. Thus, despite the same chain clay mineral, sepiolite is superior to attapulgite presumably because sepiolite contains water in amounts larger than that of attapulgite accounting for one of the factors.

It is further important that the chain clay mineral used in the present invention has a bulk density of not larger than 0.125 g/ml. When the bulk density becomes greater than the above range, it becomes difficult to uniformly disperse the particles of chain clay mineral and, hence, to obtain thixotropy maintaining stability.

In the present invention, further, it is also important that the chain clay mineral particles has a

particle size distribution of secondary particles as measured by a laser method, in which particle sizes of larger than 1.0 μm but not larger than 30 μm are not less than 70% by weight and particle sizes of larger than 1.0 μm are in a range of 5 to 30% by weight. That is, particles having particle sizes of larger than 1.0 μm but not larger than 30 μm contribute to increasing the viscosity of the dispersion solution while particles having particle sizes of not larger than 1.0 μm contribute to decreasing the viscosity. By containing these particles at a suitable ratio, therefore, the viscosity can be lowered under a condition of a large shearing force and a high viscosity can be exhibited under a condition of a small shearing force. When the contents of the above particles are not within the above ranges, the thixotropy drops. It is further desired that there are not substantially contained the particles having particle sizes of not smaller than 50 μm.

Further, the chain clay mineral that is used should have an average aspect ratio in a range of 7.5 to 9.5 and, particularly, 7.5 to 9.0. It is, further, important that the primary particles have a fibrous shape and should have an average fiber length in a range of 0.45 to 0.80 μm and, particularly, 0.50 to 0.80 μm as measured by using an electron microscope. That is, the particles having a long fiber length tend to increase the viscosity of the dispersion solution while the particles having a short fiber length tend to lower the viscosity of the dispersion solution. It is therefore necessary that the average aspect ratio and the fiber length are maintained within the above ranges to maintain a predetermined balance. A satisfactory thixotropy is not exhibited when the average aspect ratio and the fiber length lie outside

8

the above range.

It is further desired that the chain clay mineral used in the present invention contains impurity components in small amounts and, particularly, contains dolomite and calcite in small amounts. Referring, for example, to sepiolite, it is desired that a peak stemming from the dolomite near $2\theta = 31$ degrees and a peak stemming from calcite near $2\theta = 29$ degrees have the intensity ratios of peaks of not larger than 25% when a peak height stemming from the plane (110) of sepiolite is regarded to be 100% in the X-ray diffraction measurement.

Reference should be made of Fig. 1 showing X-ray diffraction images of sepiolite. Fig. 1 shows X-ray diffraction images of three kinds of sepiolite, wherein Example 1 is that of a produce of Spain containing dolomite and calcite in small amounts, the intensity ratios of peaks (2θ = near 31 degrees, 2θ = near 29 degrees) stemming from dolomite and calcite being not larger than 25% with respect to the peak (2θ = near 7 degrees) stemming from the plane (110) of sepiolite. On the other hand, Comparative Example 3 is that of Spain (different region from that of Example 1) and Comparative Example 4 is that of South Africa containing dolomite and calcite in large amounts, and the intensity ratios of peaks stemming from dolomite and calcite with respect to the peak stemming from the plane (110) of sepiolite being higher than those of Example 1. That is, as will be understood from the experimental results of Examples and Comparative Examples that will be described later, Comparative Examples 3 and 4 containing dolomite and calcite in large amounts exhibit thixotropy indexes (TI) lower than those of Example 1 containing these compounds in small amounts making it difficult to

obtain thixotropy as desired.

In the present invention, the chain clay mineral particles are obtained by using the raw material produced from a region where dolomite and calcite are contained in small amounts, finely pulverizing the raw material, and classifying them into predetermined particle sizes. As the pulverizer, there can be used a ball mill, an air pulverizer, a vibration mill, a colloidal mill, a hammer mill or a dyno mill. Among them, it is particularly desired to use an air pulverizer as represented by a jet mill. The pulverization may be carried out by a so-called wet method in the presence of a small amount of water or may be carried out by a dry method without using water. The pulverizing conditions are so set that there can be obtained the above-mentioned average aspect ratio, bulk density, primary particle shape and particle size distribution of the secondary particles. If severe pulverizing conditions are set, for example, the bulk density decreases and the fiber lengths of primary particles are shortened. Therefore, the pulverizing conditions may be suitably set depending upon the properties of the chain clay mineral that is used as the raw material.

Depending upon the region of production, further, sepiolite may contain dolomite or calcite in large amounts. In that case, dolomite and calcite are removed by the treatment with an acid such as hydrochloric acid or nitric acid, so that the intensity ratios of peaks of dolomite and calcite are adjusted to lie in the above-mentioned ranges in the X-ray diffraction.

Attapulgite contains calcite in large amounts making it difficult to satisfy the thixotropic index (TI). In the same manner as above, however, the

content of calcite can be decreased by the treatment
with an acid such as hydrochloric acid or nitric acid
to adjust the intensity ratio of peak stemming from
calcite to be not larger than 50% and, preferably, not

5   larger than 30% with respect to the peak stemming from
the surface (110) of attapulgite in the X-ray
diffraction measurement, and the mechanical treatment
is carried out such that the bulk density, average
aspect ratio and primary particle size fall within the

10   above-mentioned ranges and that the thixotropic index
(TI) lies within the above-mentioned range to express
the thixotropy (see Example 3 and Comparative Example
5).
(Thixotropy-imparting agent)

15   As described above, the thixotropy-imparting
agent of the invention comprising the chain clay
mineral particles is capable of imparting a high
degree of thixotropy when dispersed in either the
water or an organic medium, and can be effectively

20   used in the fields of coating materials, adhesives,
inks and the like by being blended in the coating
materials or adhesives that have been known per se. so
as to be used in the form of coating compositions and
adhesive compositions.

25   Examples of the coating material include
nitrocellulose coating material, alkyd resin coating
material, aminoalkyd coating material, vinyl resin
coating material, acrylic resin coating material,
epoxy resin coating material, polyester resin coating

30   material, chlorinated rubber coating material, as well
as coating materials which contain one or two or more
kinds of phenol resin, modified phenol resin, alkyd
resin, vinyl resin, petroleum resin, epoxy resin,
polyester resin, styrene resin, silicone resin,

35   chlorinated resin, urethane resin, polyamide resin,

polyimide resin, and fluorine-contained resin.

Further, the coating material that is used may be either the solvent type or the aqueous type. That is, the thixotropy-imparting agent of the invention

5   exhibits excellent properties even when dispersed in either an organic solvent or an aqueous medium.

As the organic solvent for the solvent type coating materials, there can be used toluene, xylene, n-heptane, n-hexane, cyclohexane, acetone, methyl

10   ethyl ketone, methyl isobutylketone, cyclohexanone ethanol, propanol, butanol, diacetone alcohol tetrahydrofuran, dioxane ethyl cellosolve, butyl cellosolve ethyl acetate, dimethyl sulfoxide, ethanol, methanol, ethylene glycol and glycerin in one kind or

15   in two or more kinds. Among these organic solvents, it is desired to use those having an SP value of not smaller than 10, such as alcohol solvents like cyclohexanone ethanol, propanol, butanol, ethanol, methanol, ethylene glycol or glycerin.

20   As the aqueous coating material, there can be used a coating material of the type of self emulsion or of the type emulsified with a surfactant in addition to the coating material of the type of aqueous solution. As the resin for the aqueous

25   coating material, there can be used an alkyd resin, a polyester resin, an acrylic resin, an epoxy resin or a combination of two or more kinds of them dissolved in an aqueous medium or self-emulsified. The concentration of the resin is, generally, in a range

30   of 10 to 70% by weight and, particularly, 20 to 60% by weight. The thixotropy-imparting agent of the present invention is usually blended in an amount of 0.1 to 20% by weight per the solid component.

As the adhesive, there can be used epoxy resin

35   adhesive, polyurethane adhesive, urea resin adhesive,

melamine resin adhesive, phenol resin adhesive, α-
olefin adhesive, aqueous high molecular isocyanate
adhesive, vinyl acetate resin (non-solvent type)
adhesive, vinyl acetate resin emulsion adhesive,
5   acrylic emulsion adhesive, cyanoacrylate adhesive, hot
melt adhesive, aerophobic adhesive, chloroprene rubber
adhesive, nitrile rubber adhesive, SBR adhesive,
ethylene copolymer resin adhesive, resorcin adhesive,
natural rubber adhesive, cellulose adhesive,
10   isobutene/maleic acid copolymer adhesive, alkyd resin
adhesive, furan resin adhesive, saturated or non-
saturated polyester resin adhesive, silicone resin
adhesive, polyvinyl alcohol adhesive, nylon resin
adhesive and inorganic adhesive in one kind or in two
15   or more kinds. It is desired that the thixotropy-
imparting agent of the present invention is blended in
an amount of 0.5 to 30 parts by weight per 100 parts
by weight of the adhesive resin.
(EXAMPLES)
20   The present invention will now be described by
way of Examples to which only, however, the invention
is not limited. The tests were conducted in
compliance with the methods described below.
(1) Measurement of viscosity.
25   A sample of a predetermined amount was added to a
predetermined dispersion medium (water or DOP) in a
500-ml beaker, stirred at 10000 rpm for 3 minutes by
using a household juicer to prepare about 300 g of a
dispersion solution. The dispersion solution was left
30   to stand still in a water bath at 25 °C for 30 minutes,
poured into another 500-ml beaker, and was poured
again into the initial beaker (transferred twice).
The same transfer operation was further repeated a
total of six times. By using a rotary viscometer
35   (Digital Viscometer, Model DVL-BII manufactured by

TOKIMEC Co.), the dispersion solution was stirred at a predetermined rotational speed (6 rpm or 60 rpm) for 3 minutes to measure the viscosity.

(2) Thixotropic index (TI).

A thixotropic index (TI) defined by,

$$TI = \eta_6 / \eta_{60}$$

wherein η_6 is a viscosity (at 25°C) of a dispersion solution obtained by dispersing the above chain clay mineral particles in a predetermined dispersion medium as measured at a rotational speed of 6 rpm, and η_{60} is a viscosity (at 25°C) of the above dispersion solution as measured at a rotational speed of 60 rpm, was measured by using the diethylhexyl phthalate (DOP) and water as dispersion media. When the DOP was used as the dispersion medium, the concentration of the chain clay mineral particles in the dispersion solution was 7% by weight. When water was used as the dispersion medium, the concentration of the chain clay mineral particles in the dispersion solution was 3% by weight.

(3) Bulk density.

Measured in compliance with the JIS K 6721.

(4) Particle size distribution of secondary particles as measured by a laser method.

The particle size distribution was measured by using a laser diffraction particle size analyzer, Model LS- 13 320, manufactured by Beckman Coulter Co.

(5) Average aspect ratio and average fiber length.

By using a scanning electron microscope, S-570, manufactured by Hitachi, Ltd., an average aspect ratio and an average fiber length were arithmetically calculated from five representative particles in an image of a limited visual field.

(6) X-Ray diffraction

Measured with Cu-Kα by using a Geiger-Flex RAD-IB System manufactured by Rigaku Denki Co.

Target	Cu
Filter	curved crystalline graphite monochrometer
Detector	SC
Voltage	40 KVP
Current	20 mA
Count full scale	700 c/s
Smoothing point	25
Scanning speed	2°/min
Step sampling	0.02°
Slit	DS 1° RS 0.15 mm SS 1°
Glancing angle	6°

In Examples and Comparative Examples, there were used those sepiolite and attapulgite shown in Table 1. Other properties were as shown in Table 2, and the X-ray diffraction images were as shown in Figs. 1 and 5.

Table 1

	Sepiolite A	Sepiolite B	Sepiolite C	Attapulgite A	Attapulgite B
Region of production	Spain	Spain*1	South Africa	America	India
Intensity ratio of peaks*2 (%)					
$2\theta=31°$ (dolomite)	2.0	63.7	1.0	20.4	501.1
$2\theta=29°$ (calcite)	6.3	6.3	29.4	27.1	97

*1: Region different from the region of sepiolite A.

*2: Intensity ratios of peaks of when the peak height stemming
    from the planes (110) of the samples are regarded to be 100%.

(Example 1)

The sepiolite A was coarsely pulverized, pulverized by using a roller mill, and was finely dry-pulverized by using a jet mill (PJM-100SP manufactured by Nihon Pneumatic Kogyo Co.) to obtain a thixotropy-imparting agent. The pulverizing conditions were a pulverizing pressure of 0.65 MPa and a charging amount of 2 kg/hr (the same device and conditions were also employed for the jet mill pulverization in the following Examples and Comparative Examples). The obtained thixotropy-imparting agent was measured for its properties. The results were as shown in Table 2, the X-ray diffraction image was as shown in Fig. 1, the particle size distribution of secondary particles was as shown in Fig. 2, and the SEM photograph was as shown in Fig. 3.

(Comparative Example 1)

The sepiolite A was coasely pulverized, pulverized by using the roller mill and was measured for its properties. The results were as shown in Table 2.

(Example 2)

The attapulgite A was coarsely pulverized, pulverized by using the roller mill, and was finely dry-pulverized by using the jet mill to obtain a thixotropy-imparting agent which was, then, measured for its properties. The results were as shown in Table 2.

(Comparative Example 2)

The attapulgite A was coarsely pulverized and was pulverized by using the roller mill. The pulverized product was measured for its properties. The results were as shown in Table 2.

(Comparative Example 3)

The sepiolite B was coarsely pulverized,

pulverized by using the roller mill, and was finely
dry-pulverized by using the jet mill. The pulverized
product was measured for its properties. The results
were as shown in Table 2 and the X-ray diffraction
5   image was as shown in Fig. 1.
(Comparative Example 4)

      The sepiolite C was coarsely pulverized,
pulverized by using the roller mill, and was finely
dry-pulverized by using the jet mill. The pulverized
10   product was measured for its properties. The results
were as shown in Table 2, the X-ray diffraction image
was as shown in Fig. 1 and the SEM photograph was as
shown in Fig. 4.
(Example 3)

15   The attapulgite B was coasely pulverized and was
pulverized by using the roller mill. 278 Grams of the
pulverized product was dispersed in 2500 ml of water,
300 ml of hydrochloric acid of a concentration of 3%
was added dropwise thereto, the temperature was
20   elevated to 60°C, and the treatment was conducted with
an acid for three hours (the pH of the slurry after
the treatment was 5.75). The product treated with the
acid was filtered, washed with water, dried at 110°C,
coarsely pulverized, and was finely dry-pulverized by
25   using the jet mill to obtain a thixotropy-imparting
agent. The obtained thixotropy-imparting agent was
measured for its properties. The results were as
shown in Table 2 and the X-ray diffraction image was
as shown in Fig. 5.
30   (Comparative Example 5)

      The attapulgite B was coarsely pulverized,
pulverized by using the roller mill and was measured
for its properties. The results were as shown in
Table 2 and the X-ray diffraction image was as shown
35   in Fig. 5.

Table 2

	Ex. 1	Comp. Ex. 1	Ex. 2	Comp. Ex. 2	Comp. Ex. 3	Comp. Ex. 4	Ex. 3	Comp. Ex. 5
TI (solvent: DOP[*1])								
η_6 (mPa·s)	7330	200	4120	(A)	2470	1780	7910	735
η_{60} (mPa·s)	1366	140.6	896	(A)	562	461	1571	249.2
TI	5.4	1.4	4.6	(A)	4.4	3.9	5.0	3.0
TI (solvent: water)								
η_6 (mPa·s)	562.0	817	202	883	39	8	64	60
η_{60} (mPa·s)	65.3	175	52.7	225.7	7.2	1.1	9.9	13.5
TI	8.6	4.7	3.8	3.9	5.4	7.3	6.5	4.4
Bulk density (g/ml)	0.100	0.450	0.109	0.570	0.128	0.172	0.068	0.434
Ptcl. size distribution (wt%)								
1.0 < ptcl. Diameter(μm) ≦ 30	88.0	90.9	73.3	80.1	96.0	83.7	77.2	45.2
ptcl. Diameter(μm) ≦ 1.0	11.1	0	27.6	13.0	3.9	16.3	22.4	0.1
Ave. aspect ratio (-)	8.2	14.2	8.4	13.9	7.4	5.2	8.7	24.3
Ave. fiber length (μm)	0.69	1.25	0.52	1.06	0.46	0.27	0.72	20.2
Intensity ratios of peaks[*3] (%)								
$2\theta=31$ (dolomite)	2.0	2.0	-	-	63.7	1.0	-	-
$2\theta=29$ (calcite)	6.3	6.3	27.1	27.1	6.3	29.4	21.8	97

(A): poorly dispersed[*2]
*1: DOP (diethylhexyl phthalate)
*2: Poorly dispersed due to sedimentation of the sample.
*3: Intensity ratios of peaks of when the peaks stemming from the planes (110) of the samples were regarded to be 100%.